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Local Pairing and Antiferromagnetism in High-T_c Superconductors

by

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INTRODUCTION

The CuO_2 planes in high-temperature superconductors are studied by a two-dimensional Hubbard Hamiltonian with a new proposed vacuum (d^9p^6). The superexchange between Cu sites and the hopping parameters of the carriers estimated in this new vacuum has good agreement with the experimental observation as well as numerical calculations with a finite cluster. Single carriers (electrons for n-type or holes for p-type) are localized due to the rigidity of the antiferromagnetic background. Local pairing in the CuO_2 plane enables the paired states to move around without disturbing the antiferromagnetic background. The binding energy is estimated from the pair hopping energy, which causes the paired state to be lower in energy than the single carrier state.

BACKGROUND

Since the discovery of p-type high-temperature superconductivity in ceramics [1], many theoretical studies [2] have been carried out on these compounds. There is a variety of experimental evidence that the explanation of the phenomenon does not lie within the traditional phonon-mediated mechanism. Neutron and Raman scattering experiments [3] show that strong antiferromagnetic correlations coexist in the superconducting state. Thus, the antiferromagntism is intimately related to superconductivity in these materials, in contrast to magnetism opposing superconductivity in conventional superconductors. Other prominant features, such as short coherent length (12 - 20Å) and the direct proportionality of the

superconducting phase transition temperature (T_c) to the carrier density observed in many experiments, strongly support the idea of local space pairing, which is very different from the ordinary BCS-type pairing caused by a mean-field excitation.

The hole superconductors $[La_{2-x}(Sr,Ba)_x CuO_4]$ [1] are obtained by partially substituting divalent barium or strontium for the trivalent lanthanum in La_2CuO_4. Meanwhile, the n-type superconductors $[Ln_{2-x}Ce_xCuO_4]$ (Ln: rare earth) [4] can be obtained as in semiconductors when a tetravalent lanthanide is partially substituted for the trivalent lanthanide in compounds of the form Ln_2CuO_4. The crystal structure of Nd_2CuO_4, the parent material for electron superconductors, is similar to that of the hole superconductors excpet that each copper atom is bonded to only four in-plane oxygen atoms, possessing no apical oxygen atoms above or below the CuO_2 plane. These two prominent features suggest that: (1) electron-hole symmetry must hold, and (2) the superconducting transition is purely a property of the two-dimensional CuO_2 sheets. The argument for electron-hole symmetry is strengthened by the report of both electron and hole superconductivity in $TlCa_{1-x}Ln_xSr_2Cu_2-O_{7+\delta}$ [5].

X-ray absorption spectra show that the charge carriers (electrons) are located on the Cu sites (Cu^{1+}) in n-type superconductors [6], while the charge carriers (holes) are located on O sites (O^{1-}) in p-type superconductors [7]. Clearly this observation is due to the presence or absence of electrons on the antibonding states of $Cu(d_{x^2-y^2})$ and $O(p_x$ or $p_y)$ orbitals. This kind of behavior is very similar to that of the impurity states in semiconductors. In this case, the choice of vacuum for the system plays a critical role for fixing the electron and hole energy levels, particularly for the two-dimensional Hubbard model.

The hole superconductors have been extensively studied with the Hubbard Hamiltonian [8]. In all papers using this model, the closed shell $Cu(d^{10})$-$O(p^6)$ was assumed to be the vacuum, that is, the other energy levels were well referenced to this closed shell orbit. However, this vacuum is not amenable to describing electron superconductivity. To explain both electron- and hole-state superconductors, we define a new vacuum, $Cu(d^9)$-$O(p^6)$, which exhibits an antiferromagnetic background on the Cu lattice of the parent, undoped materials. We shall study various parameters with the extended Hubbard Hamiltonian in this new vacuum, where the electron and hole creations can be explained as follows: the extra electron which is inserted within the CuO plane by doping with a

tetravalent lanthanide will occupy the empty antibonding state $Cu^{1+}(d^{10})$. If a divalent atom is the dopant instead of a tetravalent atom, the CuO_2 plane will lose one electron from the highest-occupied antibonding state $O^{2-}(p^6)$. Then the problem in the theory of high-temperature superconductors narrows down to the best description of the charge carriers (electron or hole) in an antiferromagnetic background, which is consistent with many experiments on high-tempertaure superconductors [9].

THEORY

We write the Hamiltonian as

$$H = t_{pd} \sum_{\langle i,j \rangle}' (d^{h\dagger}_{i\sigma} p_{j\sigma} + d^{e\dagger}_{i\sigma} p^{\dagger}_{j\sigma} + h.c.)$$

$$+ \sum_{i,\sigma} (E_d + U_d)\, d^{e\dagger}_{i\sigma} d^{e}_{i\sigma} + \sum_{i,\sigma} E_d d^{h\dagger}_{i\sigma} d^{h}_{i\sigma}$$

$$+ \sum_{\substack{j,\sigma \\ \sigma \neq \sigma'}} (E_p + U_p) p^{\dagger}_{j\sigma} p_{j\sigma} - \sum_{\substack{j \\ \sigma \neq \sigma'}} U_p n_{j\sigma} n_{j\sigma'} + \sum_{\substack{\langle i,j \rangle \\ \sigma,\sigma'}}' U_{pd} n^{e}_{i\sigma} n^{h}_{j\sigma'} \quad , \qquad (1)$$

where t_{pd} is the hopping integral between neighboring Cu and O sites, and the $d_{i\sigma}$'s and $p_{j\sigma}$'s are second-quantization operators related to the Cu sites and O sites, respectively. The superscripts h and e on the Cu site operators are related to the holes $Cu^{3+}(d^8)$ and electrons $Cu^{1+}(d^{10})$, respectively, due to the new definition of the vacuum, but the O site operators always represent the hole state and consequently possess the same parameters as in the old vacuum. The prime on a summation indicates $i \neq j$, and $\langle i,j \rangle$ indicates nearest neighbors between the Cu and O sites. E_d and E_p are the diagonal kinetic energies of the 3d and 2p orbitals, respectively. Notice the on-site Coulomb repulsion (U_d) of the Cu sites appears when an electron is created on a Cu site, while the on-site Coulomb repulsion (U_p) of the O sites is released when a hole is created on an O site instead of two holes being created. The interatomic Coulomb potential U_{pd} in the last term of the Hamiltonian is the electron-hole pair creation potential; it essentially gives the change in Coulomb attraction between neighboring ionic sites due to the creation of an electron-hole pair out of the vacuum.

The electron-hole pair can be created when an electron on an O site moves to a neighboring Cu site, i.e., $Cu^{2+}(d^9)-O^{2-}(p^6) \rightarrow Cu^{1+}(d^{10})-O^{1-}(p^5)$. This changes the valence charge configurations most significantly for later calculations. The oxygen 2p orbitals are assumed to be of the σ type, pointing towards the two neighboring positive Cu ions. The large covalent interaction between the Cu and O neighbors in this band can result in a rapid disappearance of the antiferromagnetic background of the Cu sites for a small amount of doping. This effect can be understood if we consider that for an electron superconductor an electron on a Cu site can hop to neighboring Cu sites only through the O site between them. The relevant energy parameters are given by $t_{pd} \approx 1.2$ eV, $U_p = 5 - 7$ eV, $U_d = 8 - 10$ eV, $U_{pd} \approx 1 - 2$ eV, and $\Delta E = E_d - E_p = 1 - 2$ eV [8,10].

In the absence of doping, the effective Hamiltonian can be written as

$$H_{cm} = J \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j \quad , \tag{2}$$

where the sum is over all the nearest-neighbor Cu sites, and $\vec{S}_i$ are spin-$\frac{1}{2}$ operators. Because the superexchange energy (J) involves an oxygen site, it is found by fourth-order perturbation theory:

$$J = \frac{4t_{pd}^4}{(\Delta E + U_d + U_{pd} - U_p)^2} \left[\frac{1}{U_d} + \frac{2}{2\Delta E + 2U_d + 4U_{pd} - U_p} \right] \quad . \tag{3}$$

This energy is significantly reduced by the on-site Coulomb potential U_d and interatomic Coulomb potential U_{pd}, and its value will be determined later.

Now any holes, produced by doping with divalent atoms or changing the oxygen content, will go onto the O site given the condition $U_p > \Delta E$. These oxygen holes will hop from site to site, becoming the carrier of the supercurrent. The effective hopping Hamiltonian of the single carrier hole can be written as

$$H_h = (t_2 - t_1) \sum_{\substack{\ell \neq \ell' \\ \sigma, \sigma'}} p_{\ell\sigma}^{\dagger} p_{\ell'\sigma'} + t_2 \sum_{\substack{\ell \neq \ell' \\ \sigma}} p_{\ell\sigma}^{\dagger} p_{\ell'\sigma} \quad , \tag{4}$$

where $p_{\ell\sigma}^{\dagger}(p_{\ell\sigma})$ creates(annihilates) holes of spin σ on the O sites. An oxygen site (ℓ or ℓ') is one of the four nearest neighbors for a given Cu site. The propagation of an oxygen hole by the first term exchanges its spin with the corresponding Cu, leaving behind a string of broken antiferromagnetic bonds among pairs of Cu sites. By means of the second term, an oxygen hole is localized over the four neighboring O sites surrounding a given Cu site. To second order in the Cu-O hopping parameters t_{pd}, we obtain

$$t_1 = \frac{t_{pd}^2}{\Delta E + U_d + 2U_{pd} - U_p} \tag{5}$$

and

$$t_2 = \frac{t_{pd}^2}{U_p - \Delta E} \quad . \tag{6}$$

Equation 3 and these expressions illustrate the dependence of the superexchange energy and the O hole hopping on the parameters of the original Hamiltonian and must be compared with the result obtained from the vacuum defined as the closed shell [8,11]. As we can see from the energy parameters, the hole hopping energy ($t_h = t_2 - t_1$) is greater than the superexchange energy between Cu sites, resulting in the propagation of the hole carriers to lower its energy.

In the case of electron superconductors, any electrons produced by doping with tetravalent atoms will obviously go into the lowest empty states, which is the antibonding state of a Cu site. Since the spins of the neighboring four Cu sites are opposite to the spin of the given Cu site, the propagation of an electron carrier always leaves behind a string of broken antiferromagnetic bonds between the adjacent Cu sites. Similar to the hole hopping, we can write the effective hopping Hamiltonian for the Cu site electron as

$$H_e = t_e \sum_{<i,j>} d_{i\sigma}^{e\dagger} d_{j\sigma}^e \quad , \tag{7}$$

where $<i,j>$ indicates nearest-neighbor Cu sites. To second order in the Cu-O hopping parameter t_{pd}, we have

$$t_e = \frac{t_{pd}^2}{\Delta E + U_d + 2U_{pd} - U_p} \; . \tag{8}$$

In equation 7, we considered only one channel for the effective hopping of the electron carrier, i.e., the electron carriers are assumed to hop through the $O^{1-}(p^5)$ configuration. The electron hopping will be reduced in equation 8 if we count the hopping through the $O^{3-}(3s^1)$ configuration on an O site, which is a process completely out of phase with equation 8. This calculation is now underway in our laboratory [13]. Using the middle values of the reported parameters given earlier -- $\Delta E = 1.5$ eV, $t_{pd} = 1.2$ eV, $U_d = 9.0$ eV, $U_p = 6.0$ eV, $U_{pd} = 1.5$ eV -- the carrier hopping energy (t_e and t_h) and the superexchange energy can be obtained. The estimated values are $t_e = 0.192$ eV, $t_h = t_2 - t_1 = 0.128$ eV and $J = 48$ meV. These results can be compared with the experimental values of $J = 50 \sim 110$ meV [3], $t_h \approx 0.15$ eV for Y-Ba-Cu-O and $t_h \approx 0.1$ eV for La-Sr-Cu-O [12]. The superexchange energy (J), which is also estimated in a cluster calculation (2 Cu atoms and 4 O atoms) [13], agrees with the above results.

Let us now consider one hole on an O site for hole superconductors and one electron on an Cu site for electron superconductors. As discussed earlier, the single carriers will lower their energies by hopping to their neighbors, but the propagation of these carriers leaves behind a string of broken Cu-Cu antiferromagnetic bonds. The price for doing this is 2J times the number of hopped-to sites. With its maximum kinetic energy of 0.77 eV, it can sweep over approximately seven or eight Cu sites. If the local spin relaxation is long enough compared to the time scale of the carrier motion, the carriers will be localized within seven to eight units cells in the linear direction. In this picture, the Cu-O plane will be fully covered with broken antiferromagnetic bonds for less than 4% concentration of the carriers. This is consistent with the rapid disappearance of the antiferromagnetic fluctuation for a small amount of doping [9,14].

As the concentration of the carriers is increased to twice that of the carrier concentration at which the antiferromagnetic disappears, the neighboring two carriers will lose their origins and make pairs by reducing the number of broken antiferromagnetic bonds between Cu sites. This local pairing in space is due to the rigidity of the antiferromagnetic background of the vacuum and is possible only when the carrier hopping energy is larger than the antiferromagnetic superexchange energy between Cu sites. This paired state can move around freely without breaking the

antiferromagnetic background of the vacuum and lowers its energy compared to the localized single electron.

We now consider the propagation of a pair of O site holes in an antiferromagnetic background hole superconductors as shown in figure 1. The propagation of a pair of Cu site electrons is the same as the O site hole propagation except that the carriers are located on Cu sites in electron superconductors as shown in figure 2. In this local pairing in space, we may consider two carriers which are close together in figure 1. The energy differences of the configurations (a)-(d) in figure 1 are due to the number of broken antiferromagnetic bonds and to intersite Coulomb repulsion. The superexchange energy can be slightly changed due to the presence of the carriers nearby [8,11]; however, we ignore this effect because it has little influence in the new vacuum. The paired holes in configuration (a) can hop to (d), which is degenerate with (a), via two intermediate states (b) or (c). By this hopping process, the paired holes can move freely, keeping the possible number of broken antiferromagnetic bonds to a minimum. The number of such bonds is determined by the Coulomb potential (V) between two carriers and the superexchange energy. The configuration (a) in figure 1 has two more broken antiferromagnetic bonds than the configuratin (c), but the Coulomb repulsion between carriers must be considered in the real system. Similar arguments for the one- and two-hole motion are discussed in simple models [15].

In this paper, we estimate the energy gap (Δ) using V as a parameter, since it is very difficult to calculate it in a microscopic domain. The Coulomb repulsions in (a) or (d) will be very small if the dielectric limit is already reached in those configurations. If this is not the case, the paired electrons will have two more broken antiferromagnets between them, but the general descriptions will not be changed. As in the case of single-carrier hopping, the effective hopping of a paired hole can be obtained by degenerate perturbation theory,

$$t_p^{(b)} = \frac{t_e^2}{2J} \tag{9a}$$

and

$$t_p^{(c)} \simeq \frac{t_e^2}{V - 2J} , \tag{9b}$$
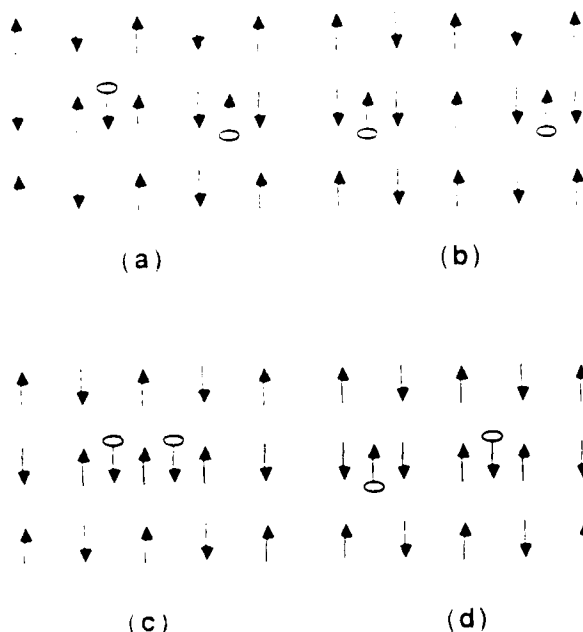
(a)   (b)   (a)   (b)

(c)   (d)   (c)   (d)

Fig. 1. Paired holes and their propagation: (a) initial state, (b) intermediate state, (c) intermediate state and (d) final state. The carrier holes are located on the O sites.
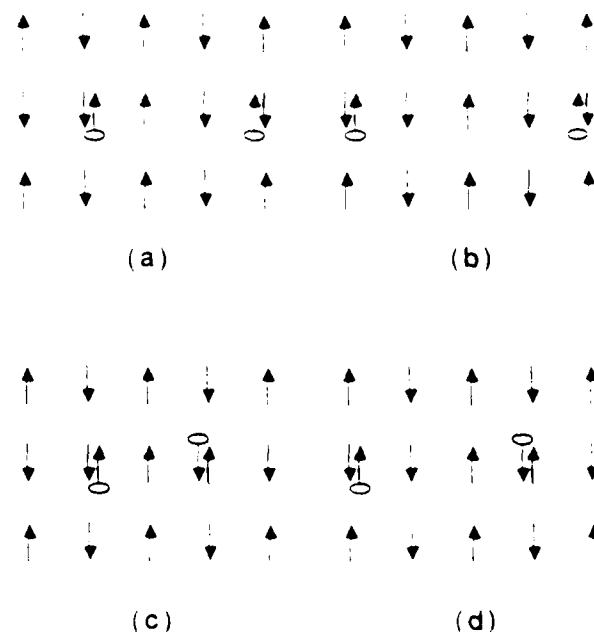
Fig. 2. Paired electrons and their propagation: (a) initial state, (b) intermediate state, (c) intermediate state and (d) final state. The carrier electrons are located on the Cu sites.
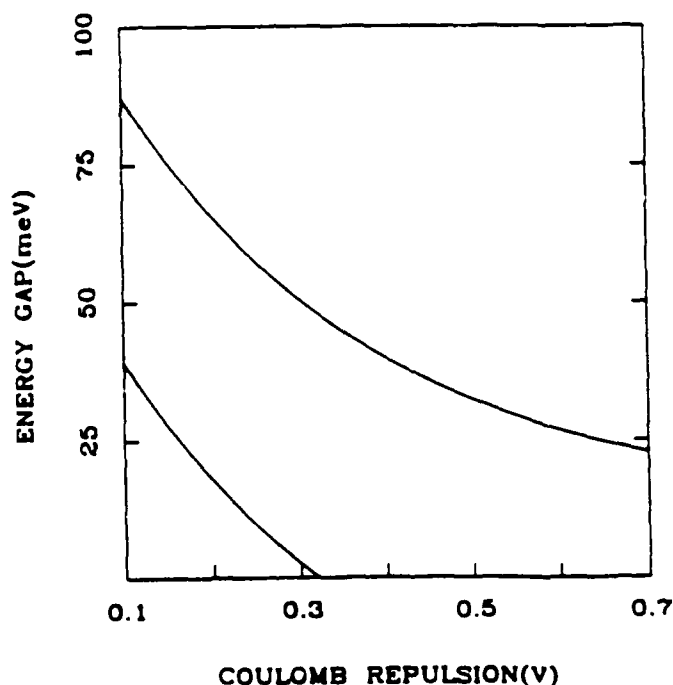

Fig. 3. Superconducting energy gap ($\Delta \sim t_p$) as a function of the Coulomb repulsion V between two carriers of the intermediate state (c) in figure 1 for the upper curve when the paired state has two spin-filled Cu sites between two carriers. V is assumed between two carriers of (a) in figure 1 for the lower curve when the paired state has three spin-flipped Cu sites between them (see the text for details).

where $t_r^{(b)}$ and $t_p^{(c)}$ are the pair hopping energy via two intermediate states (b) and (c) in figure 1, and V is the Coulomb repulsion between two carriers in (c) of figure 1. These expressions are not accurate when the energy denominators are comparable to the one hole hopping energy t_e or t_h. However, more accurate values can be obtained by a modified form of Wigner-Brillouin perturbation between three states: |initial>, |intermediate> and |final> states [8,11]. As discussed earlier, the paired holes can move freely with hopping energy $t_p^{(b)} + t_p^{(c)}$, but some of this kinetic energy is used to keep two spin-flipped Cu sites between them, which is 4J eV in energy. Therefore, the effective pair hopping energy will be $t_p - t_p^{(b)} + t_p^{(c)} - NJ$, where N is the number of spin-flipped Cu sites between two carriers. Then the energy gap, which is the energy difference between the paired state and the single-hole state, will be approximately $\Delta \sim t_p$.

NUMERICAL RESULTS AND CONCLUDING REMARKS

The energy gap versus the Coulomb repulsion of two carriers in (c) of figure 1 is shown by the upper curve in figure 3. If the Coulomb repulsion between two carriers of (a) in figure 1 is less than 0.3 eV, the paired state with three spin-flipped Cu sites between them is also lower in energy than two single carrier states. This result explains the possibility of more than one energy gap in the high-T_c ceramics, which is observed in many experiments [16]. We believe that better results can be obtained if the Coulomb interactions between valence charges in a microscopic domain, which are very difficult to estimate, are included.

Finally, we should point out that this local pairing in space is very different from the ordinary BCS-type pairing caused by a mean-field interaction. By this local pairing, all the carriers in the CuO_2 plane can make pairs and contribute to the superconducting current, which is consistent with experiments [12].

ACKNOWLEGMENTS

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